Laser-PIXE using laser-accelerated proton beams

M. Barberio, M. Scisció, S. Veltri, A. Morabito, M. Migliorati, P. Antici

Context and motivation: chemistry and physics for Cultural Heritage (CH) analysis

"Accessibility and preservation of cultural heritage is needed for the vitality of engagement within and across European cultures by also considering the importance of cultural heritage as strong economic driver in a post-industrial economy and its contribution to sustainable economic growth."

- M. Barberio, …, P. Antici: **TiO_2 and SiO_2 nanoparticles film for cultural heritage: Conservation and consolidation of ceramic artifacts**
  Surface and Coatings Technology, 271, 174 (2015)
- M. Barberio,…, P. Antici: **AFM and Pulsed Laser Ablation Methods for Cultural Heritage: Application to Archeometric Analysis of Stone Artifacts**
  Appl. Physics A 0947, 8396 (2015)
- S. Veltri, …, P. Antici: **Synthesis and Characterization of thin-transparent nanostructured films for surface protection**
  Superlattices and Microstructures 101, 209 (2017)
- M. Barberio, …, P. Antici: **Pigment darkening as case study of In-Air Plasma Induced Luminescence**
  Sciences Advances (in press)

Current Challenge in CH:

Physics and Chemistry for Cultural Heritage: obtain a complete chemical/ morphological analysis of artifacts, preventing damage

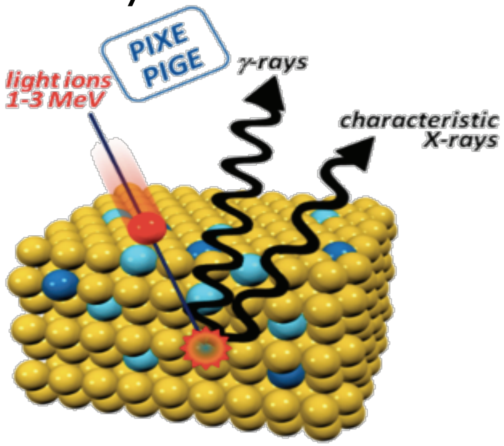
Chemical analysis:
X-ray photoelectron *Spectroscopy* (**XPS**)
X-ray fluorescence **(XRF)**
Energy Dispersive *Spectroscopy* (**EDX**)
Photoluminescence
Particle induced X-Ray Emission **(PIXE)**

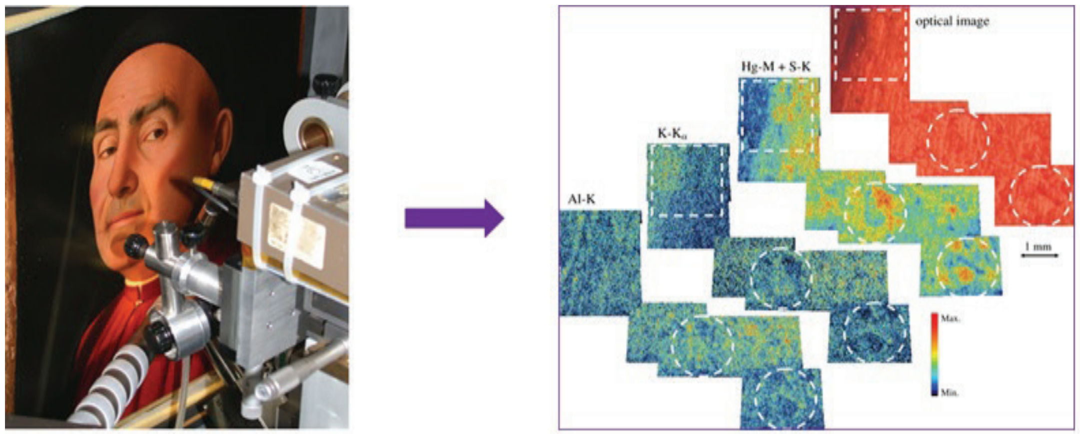
Morphological analysis:
SEM
AFM

What is Particle Induced X-ray Emission (PIXE)

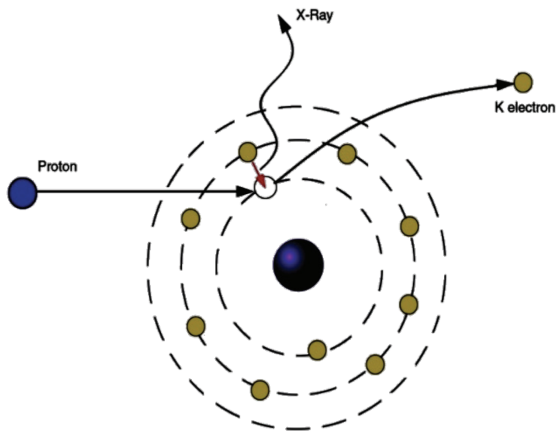
PIXE: proton beams stimulate the emission of X-rays (Gamma), which allows performing a chemical analysis of the material.

A technique used in the Cultural Heritage analysis, medicine, industry



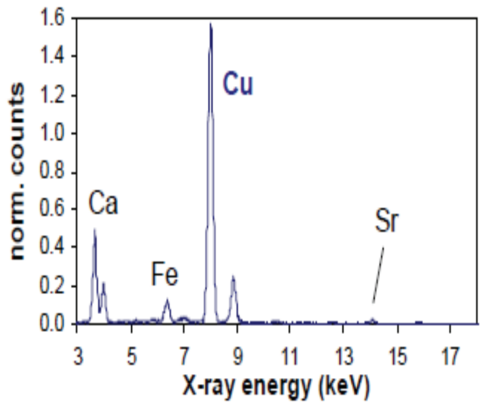


Example of PIXE to analyze the pigment's composition of The Trivulzio portrait by Antonello da Messina



K and L shell X-ray emission

- Advantage over X-Ray Fluorescence: detection of low Z elements and higher spatial precision
- Detection of elements up to 10 ppm
- Little invasive



Analysis performed at LABEC (Florence)

Current PIXE analysis with conventional accelerators

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TECHNOLOGIE
Le patrimoine du Louvre passe sous le faisceau de l'accélérateur de particules Aglaé P.14
20 minutes
www.20minutes.fr
Jeudi 23 novembre 2017 N° 3212
GRAND PARIS
ARNAQUE
Un banquier jugé pour avoir escroqué une octogénaire P.2
PRÉSIDENCE LR
La candidate Florence Portelli
Manifestation contre la réforme des retraites, en 2010.


14 Culture
Jeudi 23 novembre 2017
PATRIMOINE L'unique accélérateur de particules consacré à l'étude des chefs-d'oeuvre est au Louvre
L'art percé par le faisceau d'Aglaé
La machine permet d'obtenir la carte d'identité chimique des œuvres.
Provenance et composition
Le trésor de Bavay passé au crible
Plus de 350 bronzes
Une statuette en bronze du forum de Bavay analysée avec Aglaé.

	DR-N	
	Total	Pixel
Scan size	1 × 1 mm^2	
Pixel resolution		10 μm
Beam charge	18 μC	1.8 nC
Av. current	8 nA	
Acquisition time	35 min	210 ms
Matrix count rate	11 kc/s	
Trace count rate	69 kc/s	
Average counts		2500

A well established technique in many laboratories: AGLAE (France), LABEC (Florence), CENBG (Bordeaux) etc.

L. Pichon et al., Nucl. Instr. Methods Phys. Res. B 363, 48-54 (2015)

Laser-driven PIXE: we replace a conventional accelerator source with a laser plasma based

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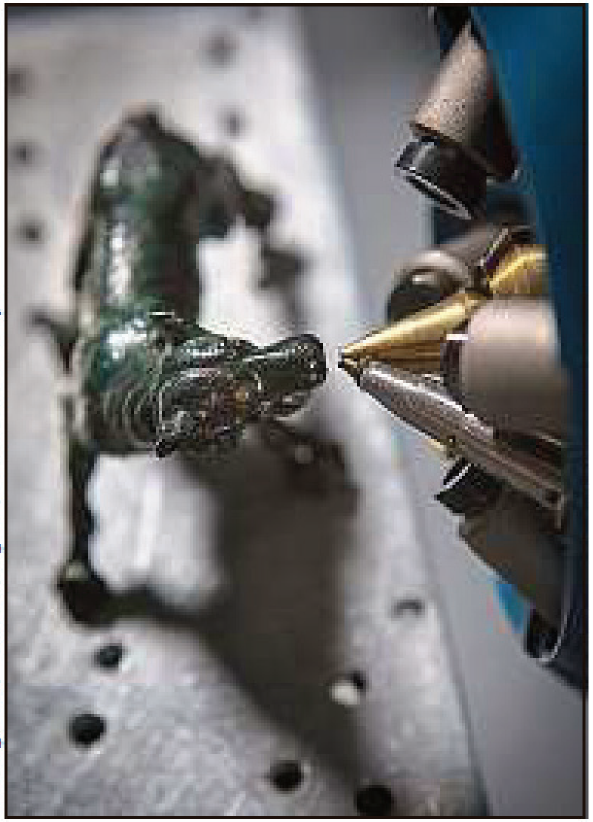
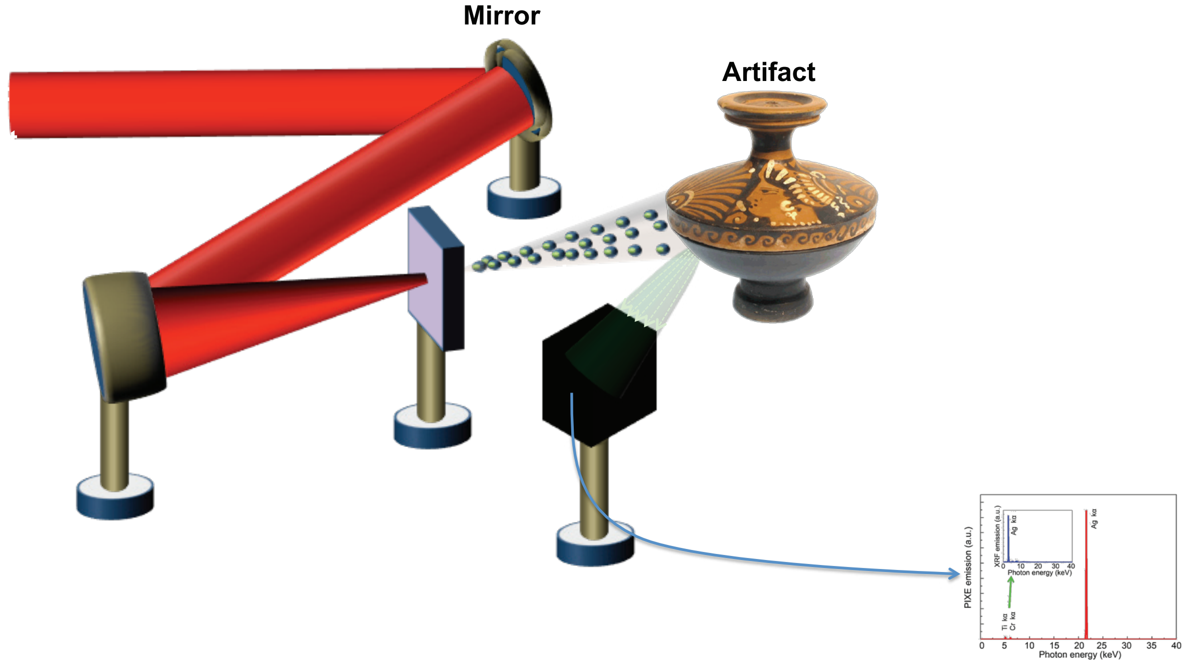


C. Hargoues/ C2RMF / Aglaé / CNRS Photothèque

Une statuette en bronze du forum de Bavay analysée avec Aglaé.



Numerical simulations to verify material sample heating (don't melt it!)

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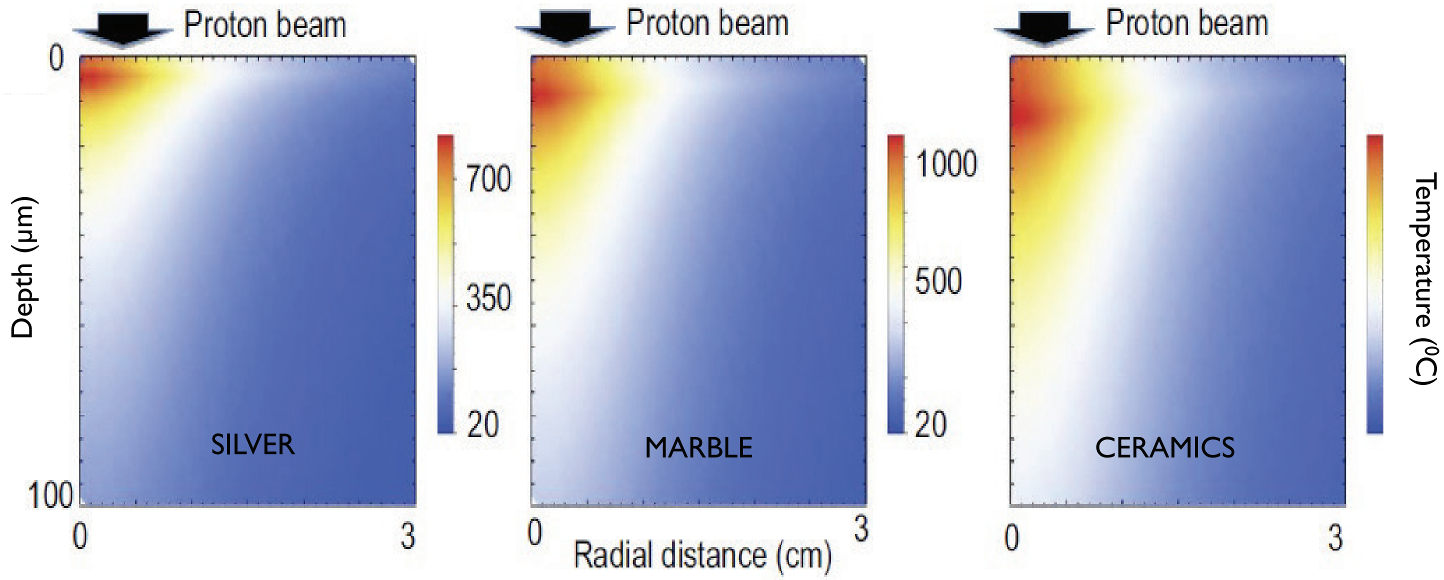
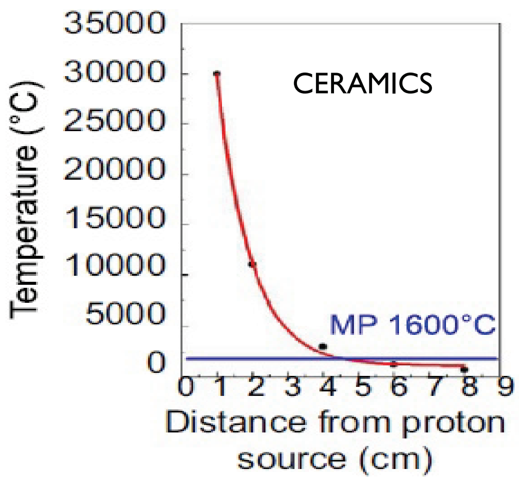
Typical CH materials:

Silver/Gold/
Bronze
Marble
Ceramics
Paper

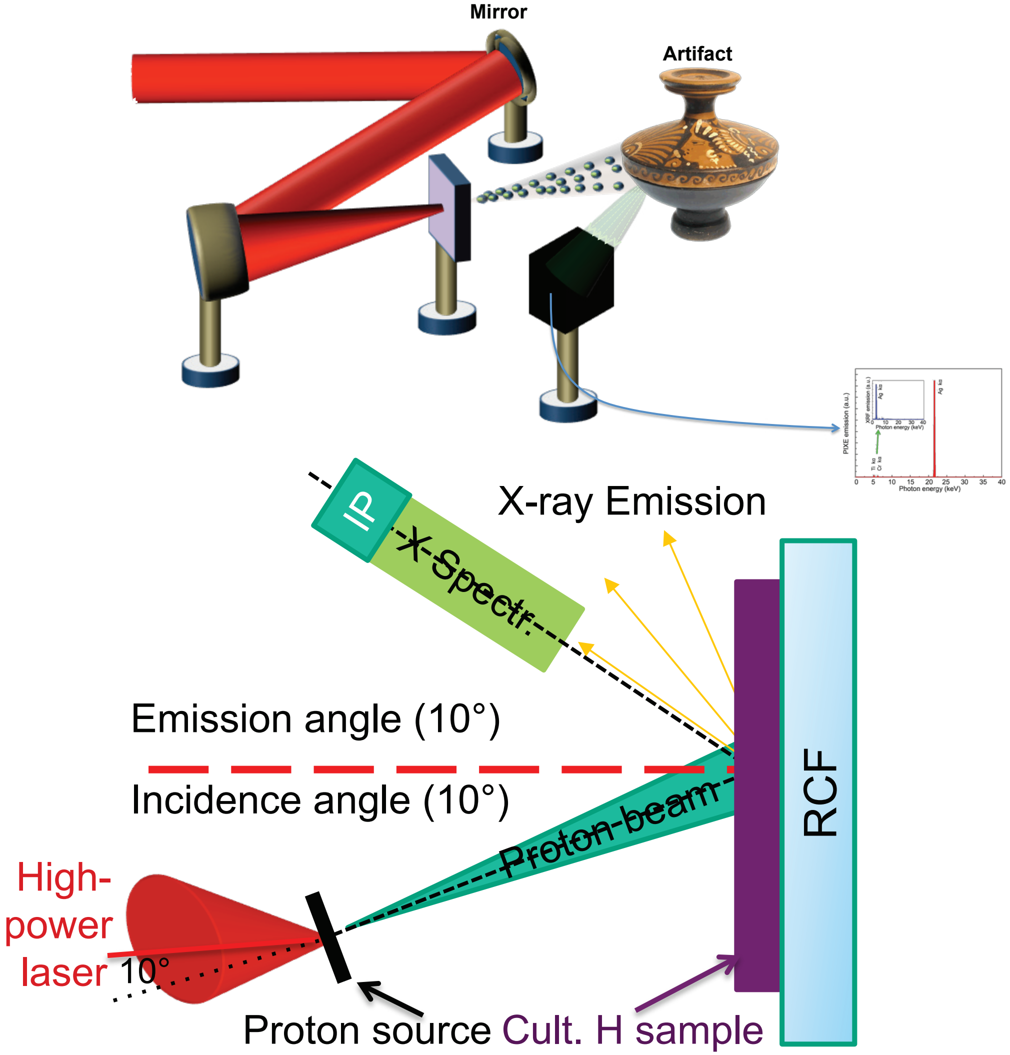


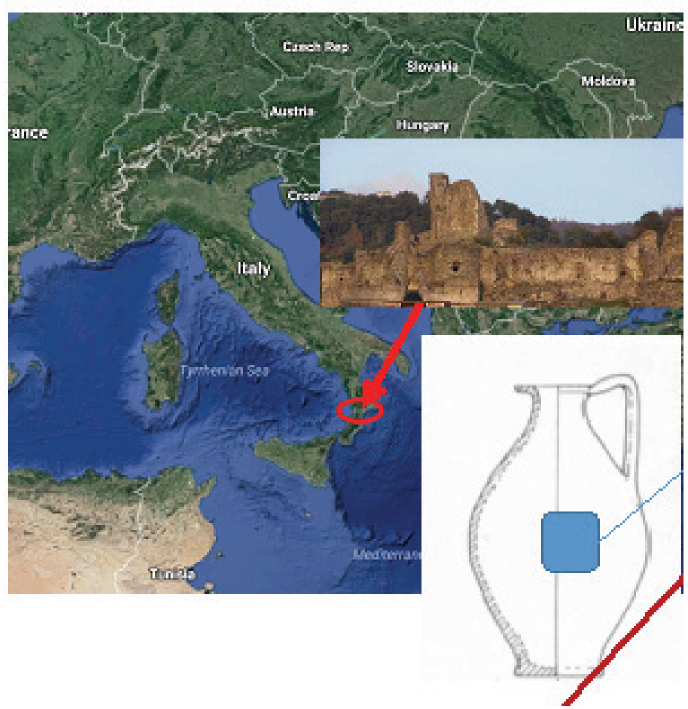
Energy deposition code simulating the TITAN spectrum (distance=6 cm)



- Temperature below meting point
- "Safe" distance in the >5 cm range
- Bunch duration in the ns range, cooling phase in the ns range

Experimental setup: first test the damage on the most sensitive material in the CH





Ceramics artifact from AD 1650 (archeological situ of Nicastro), provided by the Ministry of Culture in collabration with the Regione Calabria

Investigation of damage induced in a ceramic sample

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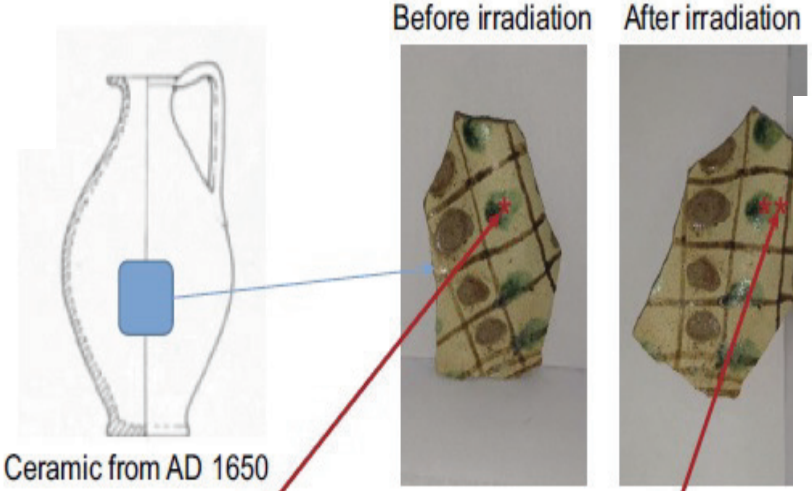
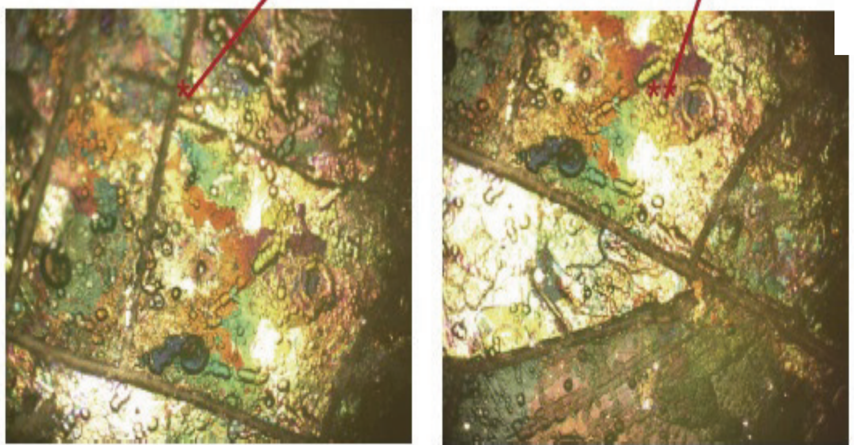
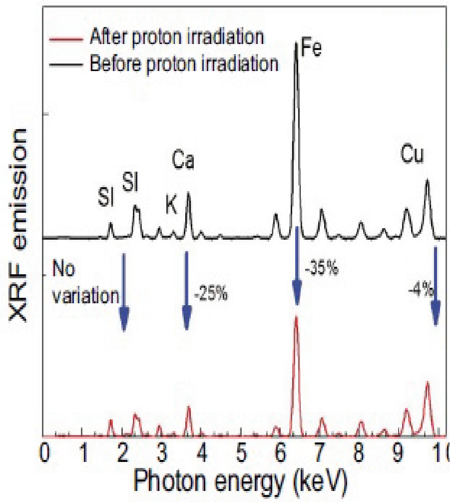


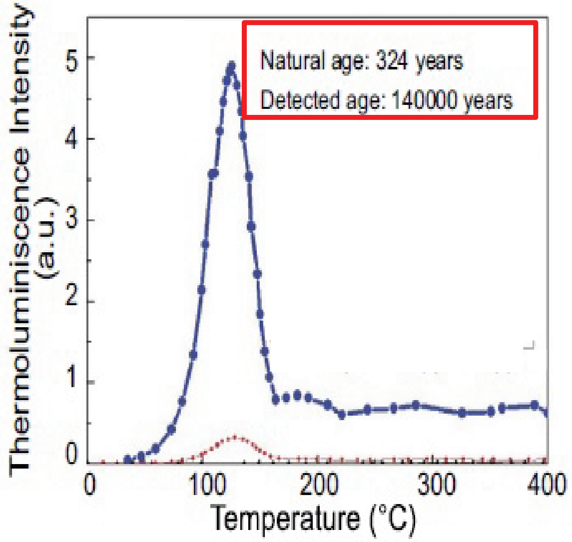
Morphological analysis: (100x mag.)



No aesthetical change after irradiation

Chemical analysis: Conventional XRF spectroscopy and Thermoluminescence





- ➢ XRF: no chemical modifications of the irradiated sample
- ➢ Absorbed dose alters the thermoluminescence of the materials: the age of the sample is artificially increased

X-ray spectrum analysis of a Silver sample with Bragg-spectrometer

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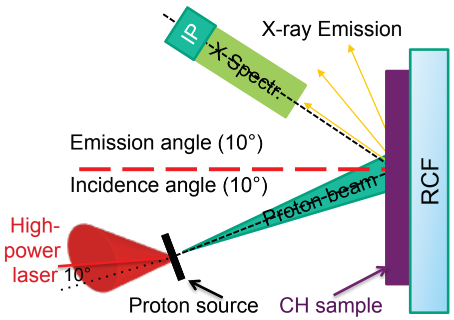
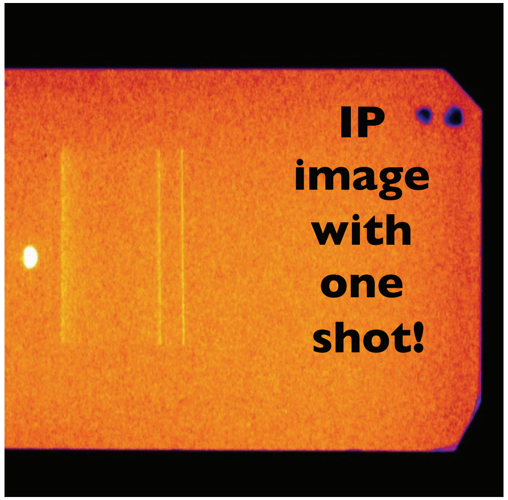
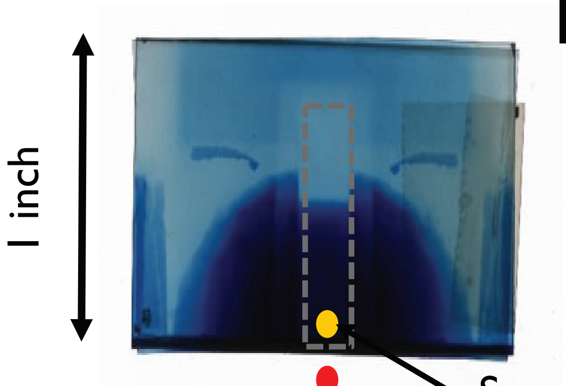






Proton beam center

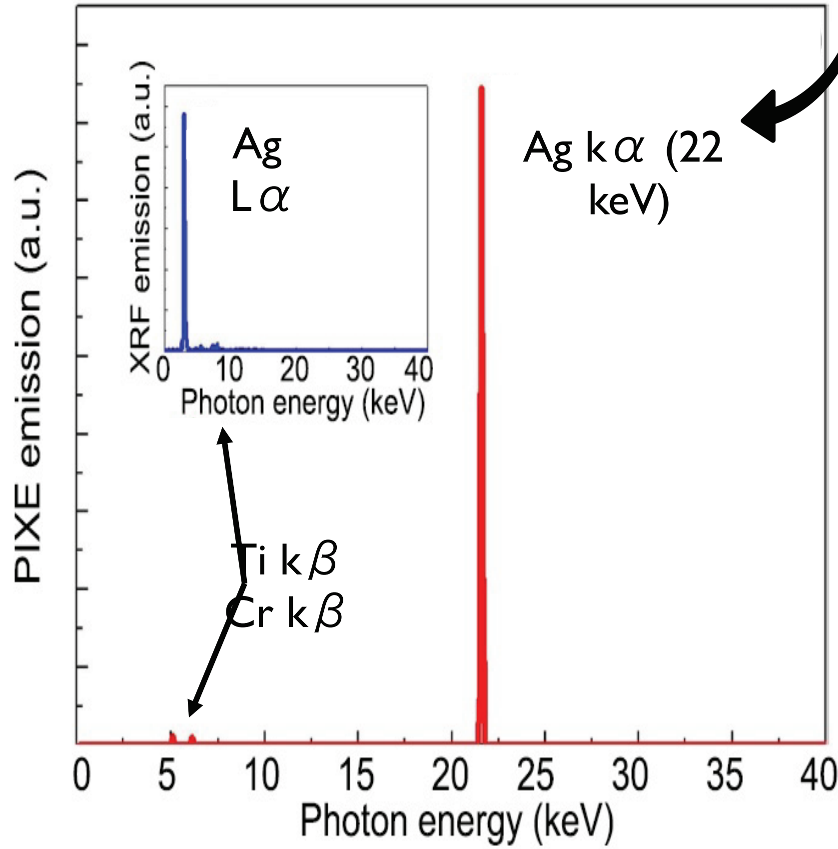
Spectrometer alignment

➢ Chemical composition of the sample successfully retrieved (Ag 97%, impurities of Ti, Cr and Cu)

Bragg's law

$$\lambda = 2d \sin\frac{\vartheta}{2}$$

$$E = \frac{hcR}{2dx}$$



Advantages and issues

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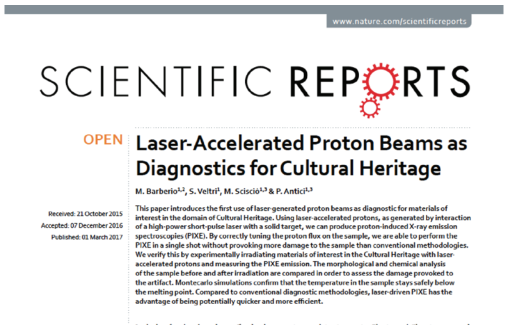

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Laser-Accelerated Proton Beams as Diagnostics for Cultural Heritage

M. Barberio[1,2], S. Veltri[1], M. Scisciò[1,3] & P. Antici[1,3]

Received: 21 October 2015
Accepted: 07 December 2016
Published: 01 March 2017

This paper introduces the first use of laser-generated proton beams as diagnostic for materials of interest in the domain of Cultural Heritage. Using laser-accelerated protons, as generated by interaction of a high-power short-pulse laser with a solid target, we can produce proton-induced X-ray emission spectroscopies (PIXE). By correctly tuning the proton flux on the sample, we are able to perform the PIXE in a single shot without provoking more damage to the sample than conventional methodologies. We verify this by experimentally irradiating materials of interest in the Cultural Heritage with laser-accelerated protons and measuring the PIXE emission. The morphological and chemical analysis of the sample before and after irradiation are compared in order to assess the damage provoked to the artifact. Montecarlo simulations confirm that the temperature in the sample stays safely below the melting point. Compared to conventional diagnostic methodologies, laser-driven PIXE has the advantage of being potentially quicker and more efficient.

M. Barberio, S. Veltri, M. Scisciò and P. Antici, Sci. Rep. 7, 40415 (2017)

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Back to results | Inventor: antici;

Method and system for analysis of objects

Abstract

A spectroscopy method and system, the method comprising irradiating an object with a laser-accelerated particle beam and detecting photons emitted by the object as a result of the interaction between the laser-accelerated particle beam and the object. The system comprises a laser; a particle source, positioned at a distance from the object; and a spectrometer and a detector; wherein the particle source generates a laser-accelerated particle beam under irradiation by the laser; and the spectrometer and the detector detect photons emitted from the object under irradiation by the laser-accelerated particle beam.

Images (7)

US20180209925A1
United States

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Inventor: Patrizio Antici, Marianna Barberio

Current Assignee : Institut National de la Recherche Scientifique (INRS) , Universita della Calabria

Worldwide applications

2018 · US CA

Application US15/877,503 events

2017-01-23 • Priority to US201762449333P

2018-01-23 • Application filed by Institut National de la

- Laser-driven proton acceleration produces routinely energies 1-11 MeV, is this useful - can you do volumetric testing ?
- What materials can be analyzed ?
- Can you go beyond 10 ppm ? How quick is the analysis ?

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ELI Beamlines and its new project: Non-Destructive Laser-driven Heritage Testing

Institute of Physics of the Academy of Sciences of the Czech Republic / E
a new project called Non-Destructive Methods of Heritage Testing , fina
(Pól růstu ČR, the City of Prague). Researchers from ELI-Beamlines work
Acceleration by Laser, and currently commissioning the ELIMAIA user be
the project by the Institute of Nuclear Physics in Řež and Istituto Nazion
Nazionali del Sud (INFN LNS) in Catania. INFN ranks among the best in
and restoration of monuments, but with a conventional particle acceler
protons will be worldwide unique for the combination of the planned a

Intense Laser Irradiation Laboratory

National Institute of Optics – National Council of Research

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Laser-PIXE

Laser driven Particle Induced X-ray Emission:
source development and X-ray spectral/spatial analysis

The PIXE (Particle Induced X-ray Emission) is a multi-elemental, quantitative, highly s
ments with concentrations in parts-per-million), rapid, non-invasive and non-destructive
to determine the composition of surface layers of a sample. The technique is based on s
characteristic X radiation emitted by each atom following irradiation with proton/ion bea
protons with MeV energies (2-3 MeV typically) that are currently produced with particle
dimensions (and costs) represent a limit for the application of the same technique outside o

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erc

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People

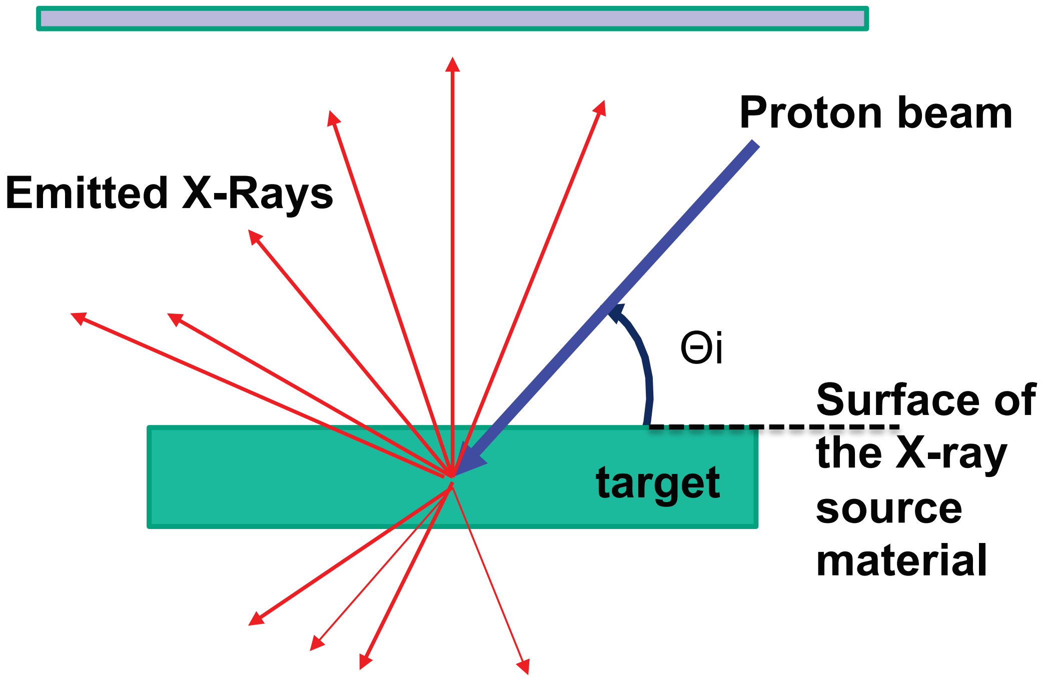
Core Team

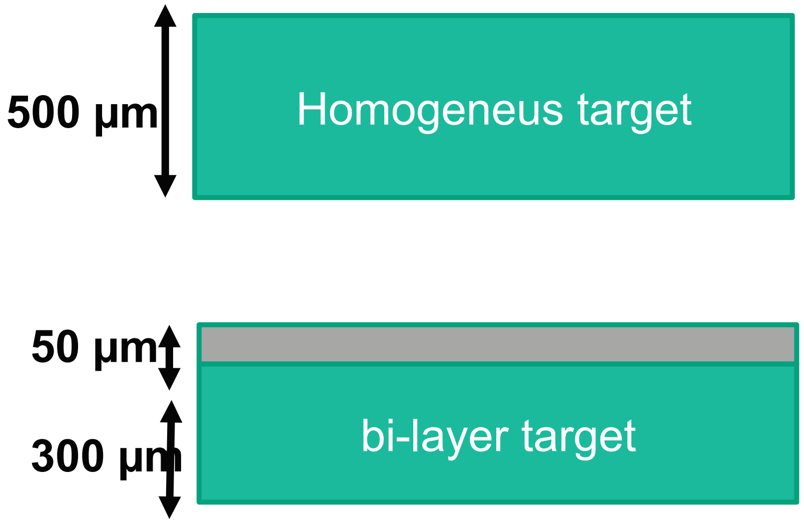
Principal Investigator

Matteo Passoni

Optimization of laser-PIXE using GEANT4

CCD DETECTOR (4 cm x 4 cm x 250 μm)
(solid angle ~0.81 str)





GEANT4 with the package G4ParticleGun, cylindrical symmetry, number of particles between 10^4 and 10^{13}. The proton beam diameter = 1 μm (typical applications)

M. Pia, et al, PIXE simulations with Geant, *IEEE Trans. Nucl. Sci.*, **56**, 3614 (2009)

Reproducing the experimental results on a silver sample

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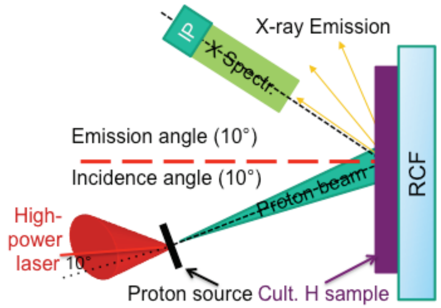
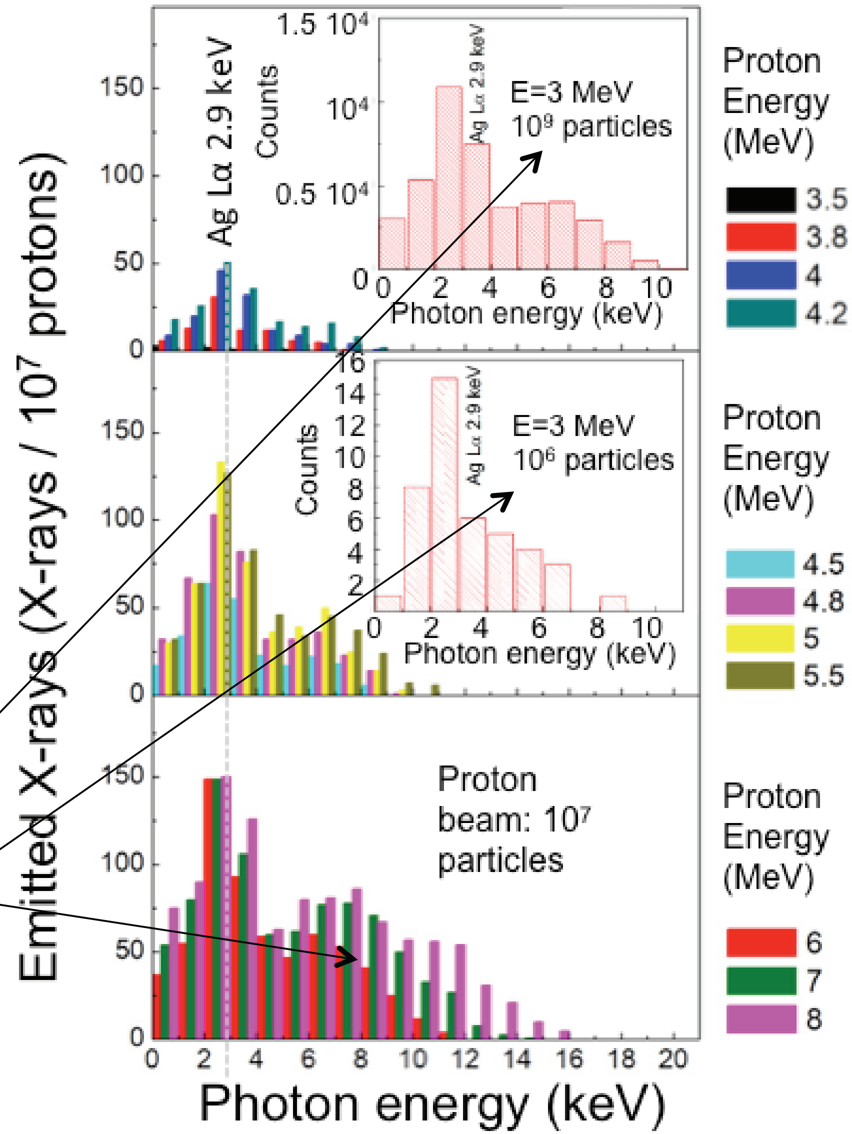


Efficiency 10^{-4} photons/proton
About 10^{13} protons/MeV/str @ 3 MeV
->10^{-9} photons / str - close to experimental value

Very high Bremsstrahlung
Energies above 5 MeV of little interest

Ratio is independent from particle number



Optimization of the incident angle: 60 degrees incident angle favors Bremsstrahlung for 3 MeV incident protons on SiO_2

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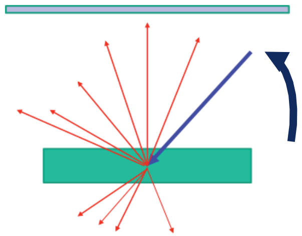
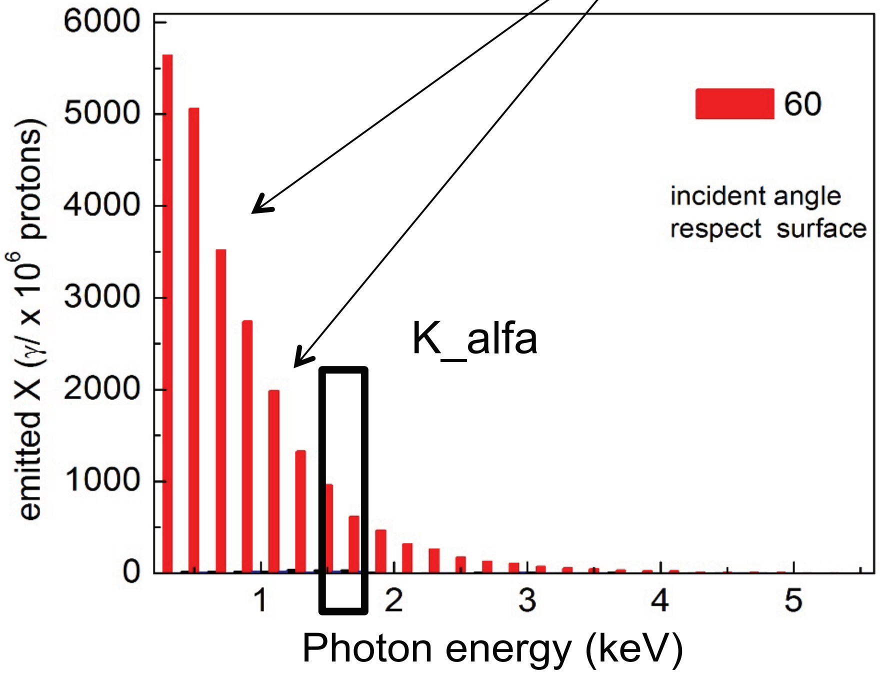
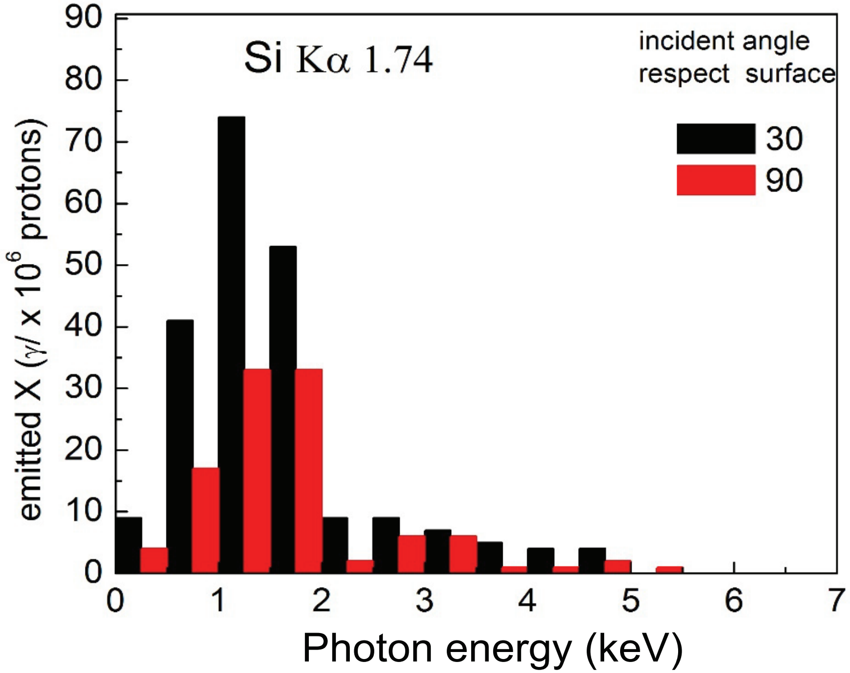


Optimized angle for the detection of the K_alfa is around 30°

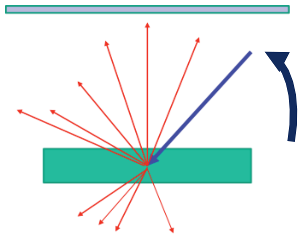
Significant Bremsstrahlung

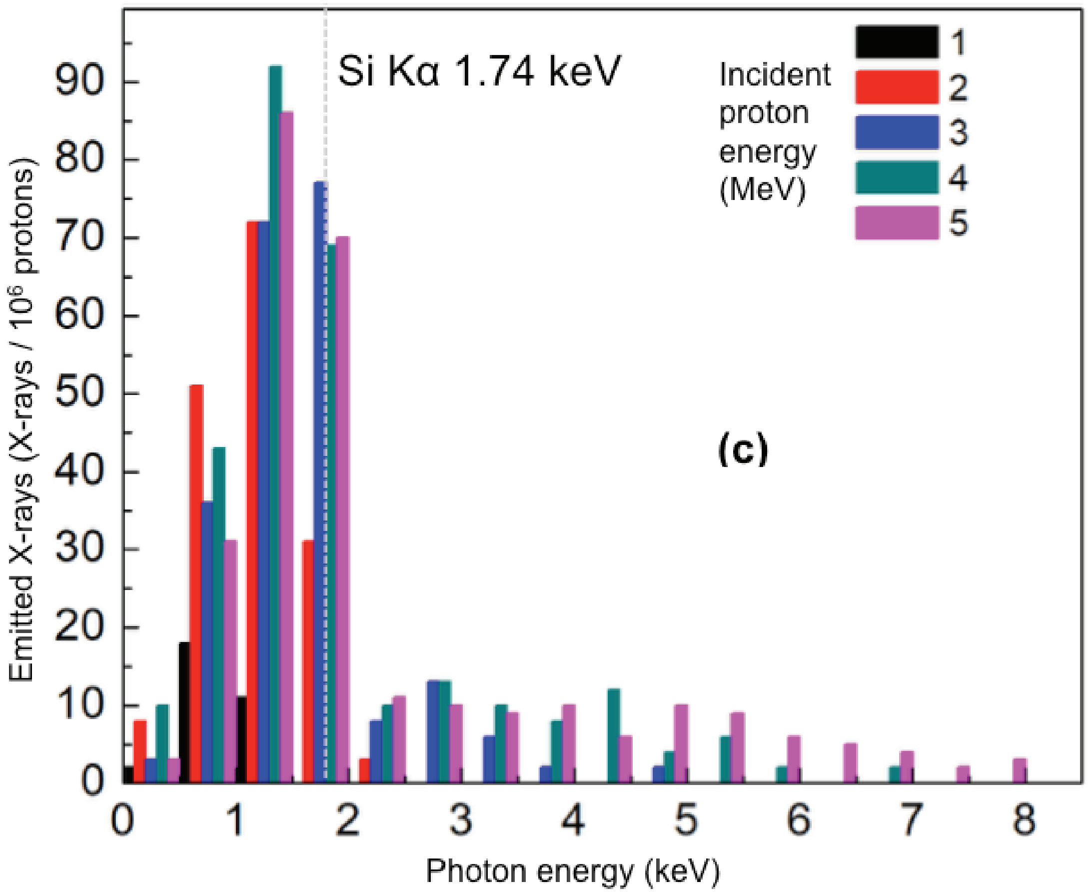




One of the most analyzed materials: Ceramics (SiO_2), Mean energy 3 MeV, 10^6 protons, 3 cm distance

30° incident angle better also for other incident energies





In the following we chose 30° as best incident angle !

One of the most analyzed materials: Ceramics (SiO_2), different mean energies, 10^6 protons, 3 cm distance

Contribution of the different depth to the total X-ray production

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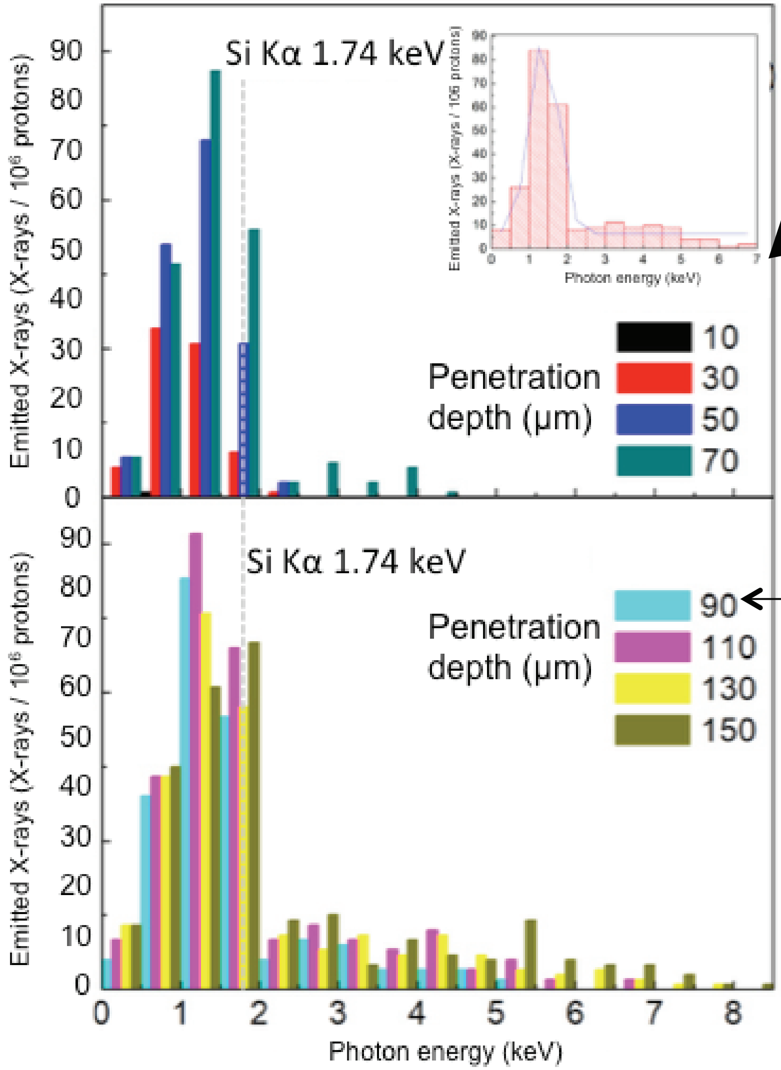
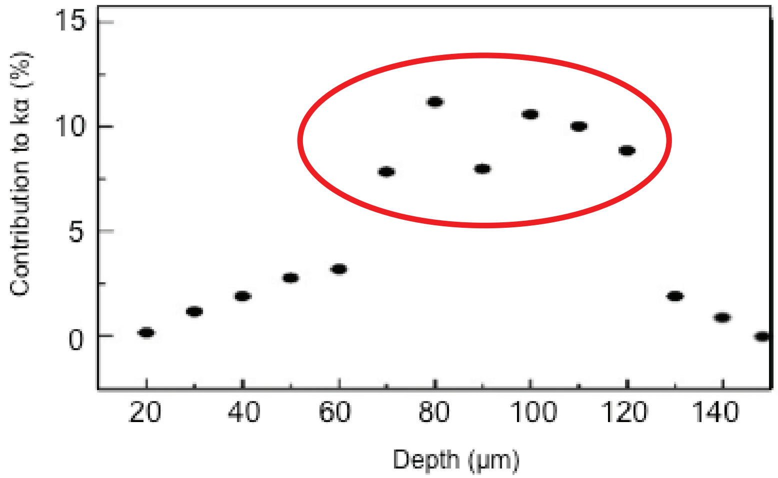


Bin size = 0.5 keV

3 MeV

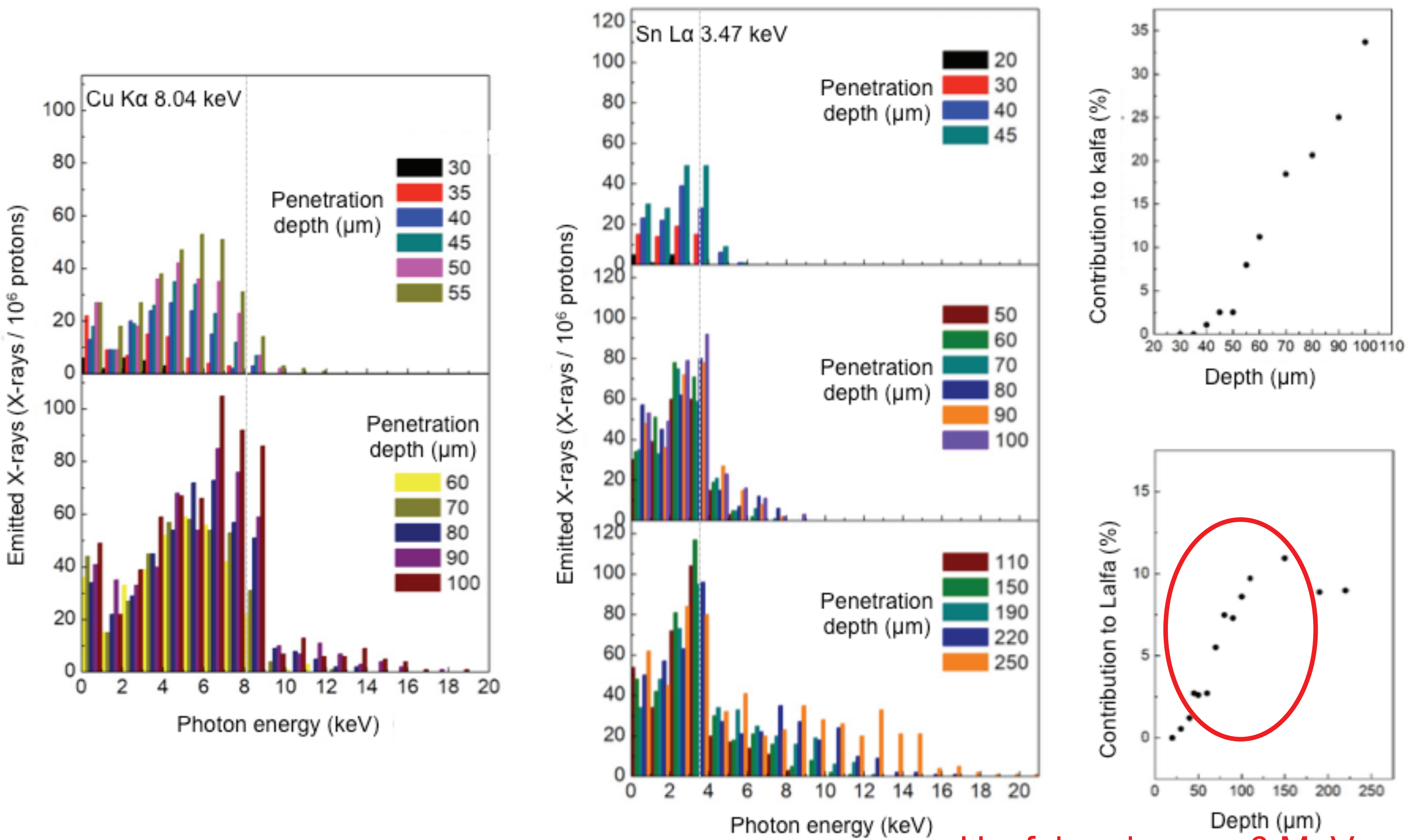
Bin size = 0.02 keV



Useful region: ~2.5 – 4 MeV

One of the most analyzed materials: Ceramics (SiO_2)

We verify this on a bronze alloy (material which consists of 90 % Cu and 10 % Sn)



Useful region: <~ 6 MeV

How does this compare with other Multi-layer targets ??

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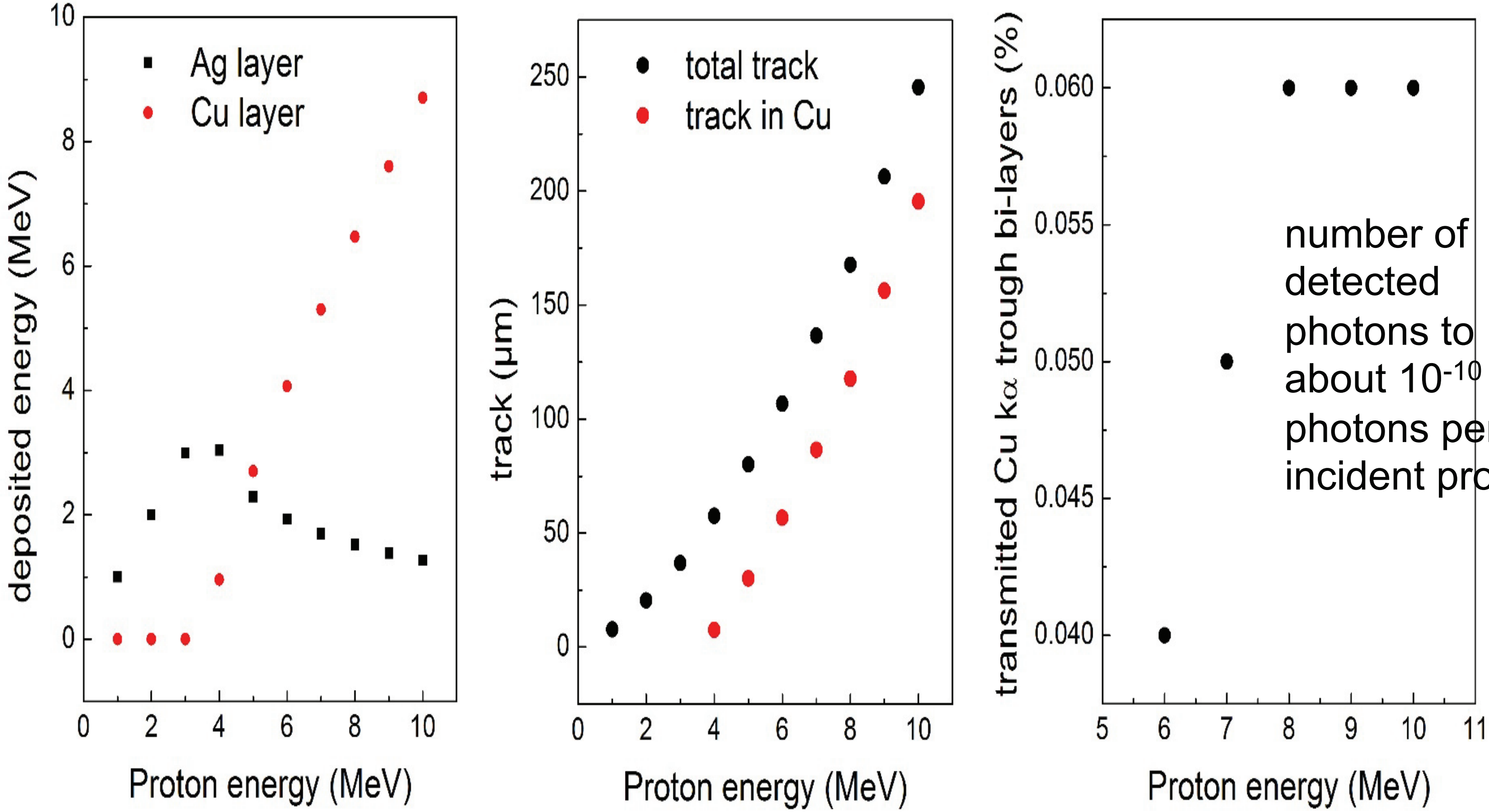
Ag(50µm)//Copper(300µm) multilayer sample



number of detected photons to about 10^{-10} photons per incident proton

For other multilayer targets ?

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- For example Pb//SiO_2 or Ti//SiO_2) the transmittance of photons generated in the SiO_2 layer is extremely low (close to zero) due to the very low energy of the SiO_2 K_alfa photons.

- Multilayers consisting of Pigments//Ceramic and Pigments//Canvas, simulated as a PbO layer on SiO_2 (or $CaCO_3$ for the canvas), or $Ti0_2$ layer on SiO_2 is the same.

Laser-PIXE very difficult for multilayers with Kα < 6 keV…

Again, we are not alone !

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Laser-PIXE using laser-accelerated proton beams

M. Barberio & P. Antici

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Laser-driven proton acceleration is a field of growing interest, in particular for its numerous applications, including in the field of materials science. A benefit of these laser-based particle sources is their potential for a relative compactness in addition to some characteristics at the source that differ from those of conventional, radio-frequency based proton sources. These features include, *e.g.*, a higher brilliance, a shorter duration, and a larger energy spread. Recently, the use of laser-accelerated protons has been proposed in the field of Cultural Heritage, as alternative source for the Particle Induced

M. Barberio, P. Antici, Sci. Rep. **9**, 6855 (2019)

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Superintense Laser-driven Ion Beam Analysis

M. Passoni, L. Fedeli & F. Mirani

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Published online: 24 June 2019

Ion beam analysis techniques are among the most powerful tools for advanced materials characterization. Despite their growing relevance in a widening number of fields, most ion beam analysis facilities still rely on the oldest accelerator technologies, with severe limitations in terms of portability and flexibility. In this work we thoroughly address the potential of superintense laser-driven proton sources for this application. We develop a complete analytical and numerical framework suitable to describe laser-driven ion beam analysis, exemplifying the approach for Proton Induced X-ray/Gamma-ray emission, a technique of widespread interest. This allows us to propose a realistic design for a compact, versatile ion beam analysis facility based on this novel concept. These results can pave the way for ground-breaking developments in the field of hadron-based advanced materials characterization.

Can we improve perform also layer-by-laser analysis and optimize the proton yield to improve the diagnostic ?

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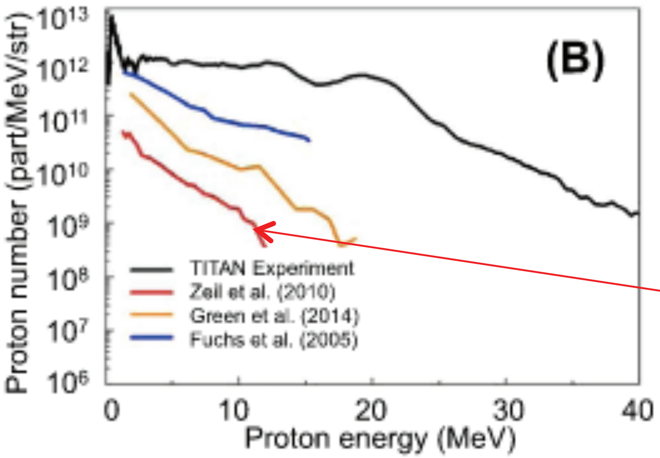
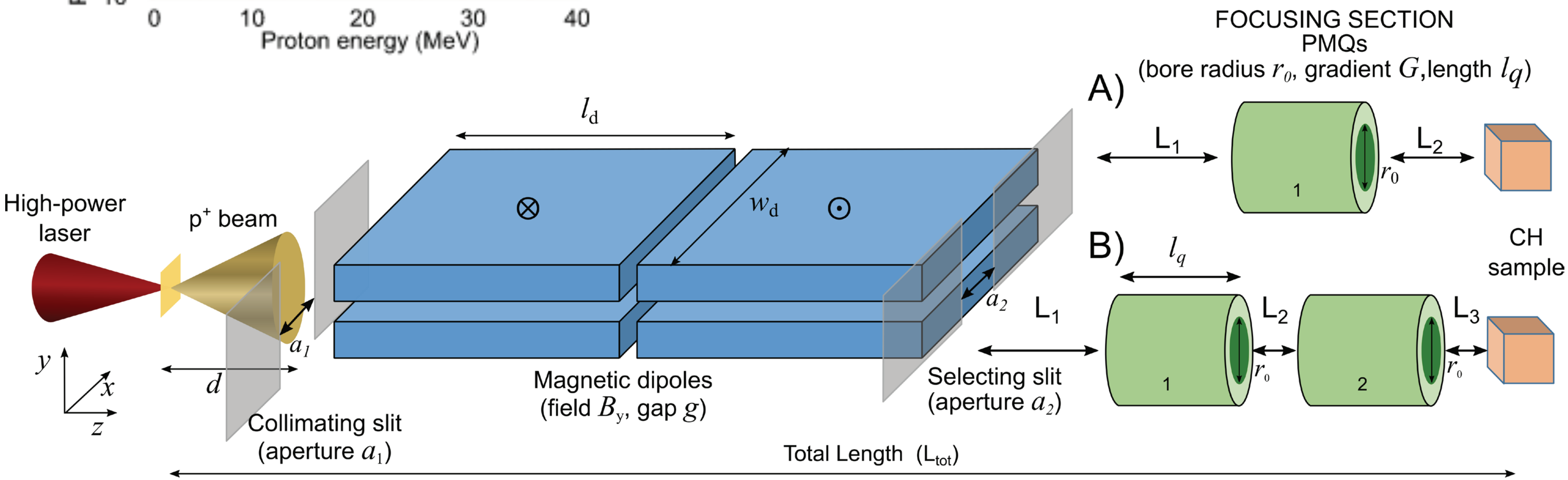
We setup a mini-beamline to be implemented on typical commercial 200 TW lasers:



Chosen proton spectrum



Particle tracing simulations for 1 PMQ

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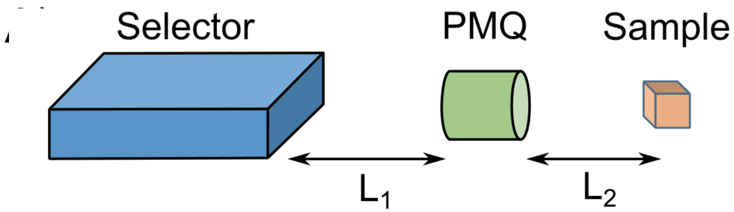
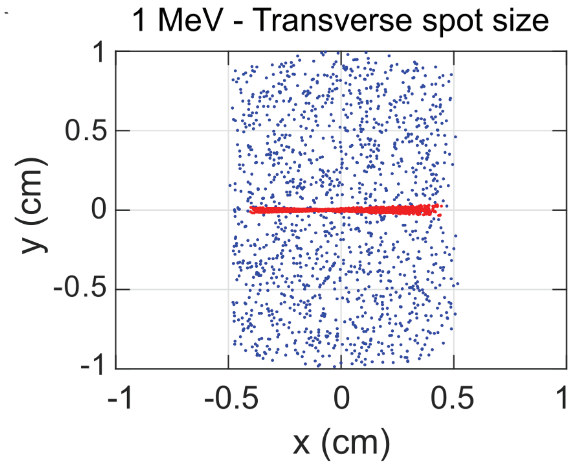
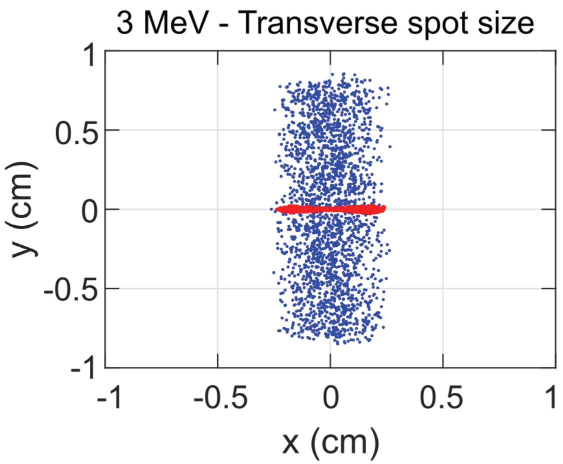
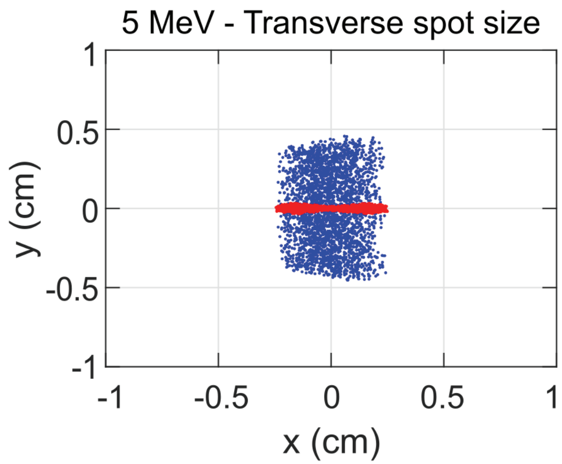


E± ΔE (MeV)	L_1 (cm)	L_2 (cm)	x (cm)	y (cm)	Q (nC)
1±0.03	31.0 - 17.0	4.5 - 18.5	0.89 - 1.10	0.05 - 1.70	0.05
3±0.12	25.7 - 3.0	9.8 - 32.5	0.55 - 0.48	0.05 - 1.20	0.02
5±0.25	21.1 - 5.0	14.4 - 30.5	0.55 - 0.60	0.06 - 0.90	0.004







Particle tracing simulations for 2 PMQ

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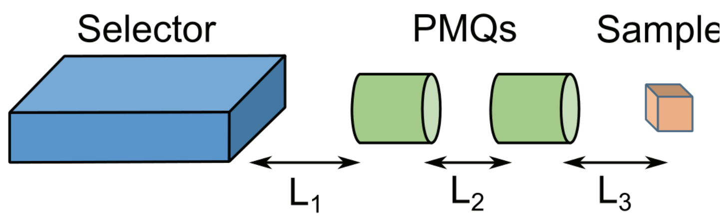
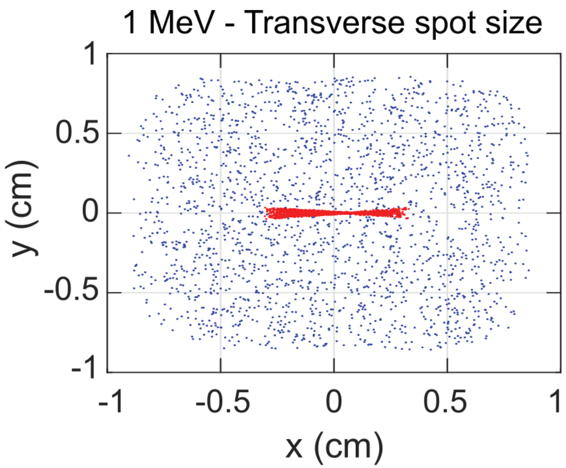


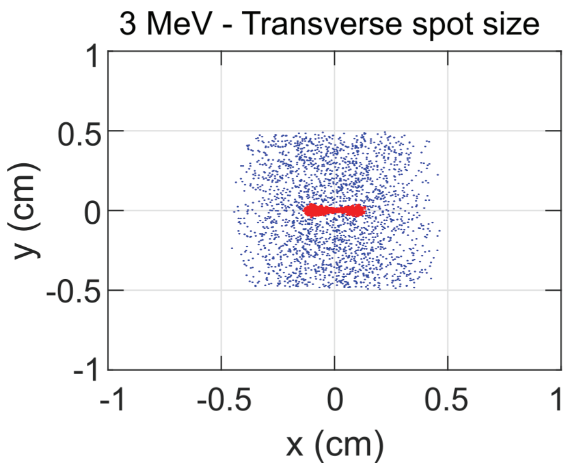
E± ΔE (MeV)	L_1 (cm)	L_2 (cm)	L_3 (cm)	x (cm)	y (cm)	Q (nC)
1±0.03	32.5 - 24.0	0.5 - 9.0	1.0 - 1.0	0.50 - 1.60	0.03 - 1.80	0.05
3±0.12	24.4 - 5.0	3.6- 23.9	6.0 - 5.1	0.20 - 0.80	0.04 - 1.00	0.02
5±0.25	9.5 - 5.0	6.2 - 24.7	18.3 - 4.3	0.10 - 0.60	0.10 - 0.60	0.004

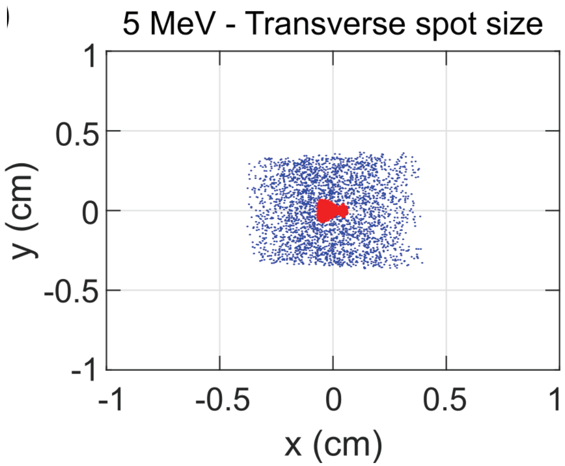


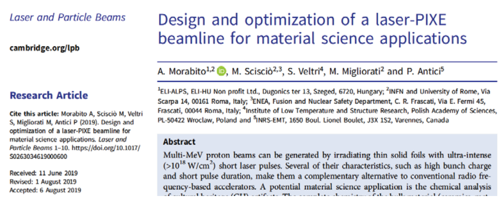

SCIENTIFIC REPORTS

OPEN Design and optimization of a compact laser-driven proton beamline

M. Scisciò et al.,
Sci. Rep. 8, 6299 (2018)






Laser and Particle Beams

cambridge.org/lpb

Research Article

Design and optimization of a laser-PIXE beamline for material science applications

Abstract

A. Morabito, M. Scisciò, S. Veltri, M. Migliorati, P. Antici (2019) Laser and Particle Beams 1–10.

Next steps

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Experiments to verify the Laser-PIXE efficiency using laser-accelerated protons using the newly commissioned ALLS Proton beamline (Nov 2019)

- Check/develop a suitable X-ray detector (0.5 – 20 keV, able to detect low photon numbers)
- Verify the experimental conditions for performing laser-PIXE
  - How many shots are needed to see a signal ?
  - Is it possible to see materials up to what depth
- Compare the damage induced to the materials by conventional PIXE and laser-PIXE
- Check the layer-by-layer analysis

Apply PIXE for Aerols (bigger advantage than Cultural Heritage ?)

Contents lists available at ScienceDirect
Nuclear Instruments and Methods in Physics Research B
journal homepage: www.elsevier.com/locate/nimb

Present role of PIXE in atmospheric aerosol research

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Ghent University, Department of Analytical Chemistry, Krijgslaan 281, S12, BE-9000 Gent, Belgium
Department of Pharmaceutical Sciences, University of Antwerp (Campus Drie Eiken), Universiteitsplein 1, BE-2610 Antwerp, Belgium

Development of a portable PIXE system for aerosol monitoring

S.Matsuyama, K.Ishii, D.Izukawa, H.Yamazaki, Ts.Amartaivan, K.Hotta, [1]S.Futatsugawa [2]K.Sera

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Coll:

Interested ?
…collaborate or join us !

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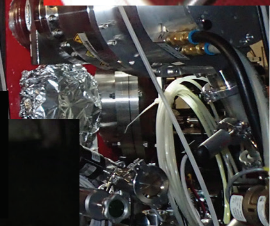

Thank you for your attention !

iPAT-LAB